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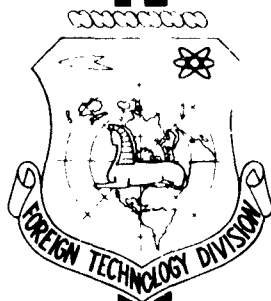
## TRANSLATION

HETEROGENEOUS COMBUSTION IN A SYSTEM  
CONTAINING CONDENSED COMPONENTS

By

N. N. Bakhman and D. P. Polikarpov

## FOREIGN TECHNOLOGY DIVISION

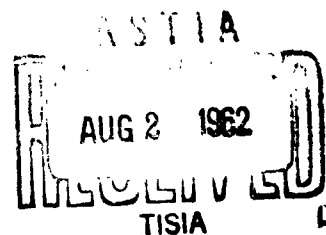


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## UNEDITED ROUGH DRAFT TRANSLATION

### HETEROGENEOUS COMBUSTION IN A SYSTEM CONTAINING CONDENSED COMPONENTS

By: N. N. Bakhman and D. P. Polikarpov

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TRANSLATION SERVICES BRANCH  
FOREIGN TECHNOLOGY DIVISION  
WP-AFB, OHIO.

HETEROGENEOUS COMBUSTION IN A SYSTEM  
CONTAINING CONDENSED COMPONENTS

N. N. Bakhman and D. P. Polikarpov

(Moscow)

The patterns of heterogeneous combustion depend considerably on the initial aggregate state of the components of the system and on their aggregate state during the course of combustion. In particular, in a case little studied, that of heterogeneous combustion, where both components are initially in the solid phase, the nature of the combustion depends to a strong extent on the relative ability of the components to form a gaseous phase during the course of combustion. It was shown previously [1] that if both components form a gaseous phase comparatively easily, there exists, firstly, a minimum scale of heterogeneity\*, below which the combustion is indistinguishable from the combustion of a homogeneous system of the same chemical composition, and secondly, for a sufficiently large scale of heterogeneity the burning velocity remains practically unchanged as the scale increases

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\* The scale of heterogeneity depends on the size of the particles of both components, as well as on the volume relationship between the components, the degree of mixing, and the relative density of the system.

further. This is due to the propagation of the flame along the surfaces of contact of the components and is very pronounced in the case of ordered systems, where the layers of the components are oriented in the direction of propagation of the flame. In this case the burning velocity is entirely determined by the velocity  $v$  of the forward points of the flame along the surface of contact of the components\*; moreover, the thickness of the burning zone increases, as the scale of heterogeneity increases.

This paper was devoted to a study of the propagation of a flame along a plane and cylindrical surface of contact of a number of solid inorganic oxidizing agents incapable of burning in their pure form ( $KClO_4$ ,  $KClO_3$ ,  $KMnO_4$ ,  $BaO_2$ ) and solid fuels: polymethyl methacrylate (Plexiglas), polyethylene, polystyrene, polyvinyl chloride (vinylplast), and others. The tests were conducted in an inert gas (nitrogen) at the pressures  $p = 0$  to 100 atm (gage).

The over-all picture of the combustion. The system was ignited with a Nichrome spiral on the upper end simultaneously along the entire length of the boundary of contact of the components. After a short initial acceleration section a sufficiently constant flame speed was usually established. In most cases the combustion takes place very energetically with the formation of a large jet of flame. A wedge-shaped cavity forms in the fuel layer and moves together with the flame. The oxidizing agents with which the work was carried out yield a condensed residue after combustion. The presence of this

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\* In the case of nonordered systems with a chaotic arrangement of particles the process of the transmission of combustion between adjacent particles of a given component is also important. It occurs as a result of the input of heat from neighboring sections of the system. If the ratio between the scale of heterogeneity and the diameter of the system is too large, the combustion can become unstable and be extinguished.

residue distorts the shape of the cavity forming in the oxidizer layer. The burning of systems containing  $KClO_4$  and  $KClO_3$  is accompanied by the formation of large drops of liquid  $KCl$ , which close off a considerable portion of the surface of the fresh oxidizer; from time to time drops are ejected by the gas stream and new ones grow in their place. The burning of systems containing  $BaO_2$  and  $KMnO_4$  is accompanied by the formation of a solid porous sediment, the volume of which is close to the volume of the initial oxidizer.

The dependence of  $v$  on the size of the oxidizer layer. Tests were conducted on the following system: a flat layer of variable thickness  $d$  of oxidizer between two thick\* layers of fuel. The flame speed was measured by a photorecorder. Figure 1 shows the curves  $v(d)$  for a layer of  $KClO_4$  and  $BaO_2$  between thick layers of Plexiglas. In both cases the curve  $v(d)$  at sufficiently great  $d$  emerges onto a plateau\*\*. However, when  $d$  decreases, the behavior of the curves  $v(d)$  is different. In the case of the system Plexiglas- $KClO_4$ -Plexiglas the flame speed increases rapidly with decreasing  $d$ . It might be expected that at sufficiently small  $d$  the value of  $v$  will pass through a maximum and begin to fall, but this effect was not observed, owing to the difficulties of obtaining thin (of the order of tenths of millimeters) layers of  $KClO_4$ . In the case of the system Plexiglas- $BaO_2$ -Plexiglas the value of  $v$  decreases smoothly with decreasing  $d$ . When  $d = 1.0$  to  $1.5$  mm, this system yields unstable results, since

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\* In comparison with the thickness of the layer burnt up during the course of the test.

\*\* In the following sections of this article (effect of nature of components, of pressure, of the relative density of the oxidizer)  $d$  was chosen large enough so as to be able to operate within the limits of the plateau on the curve  $v(d)$ .



the sediment forming during the decomposition of  $\text{BaO}_2$  begins to be ejected by the stream of gases; moreover, the flame speed increases sharply.

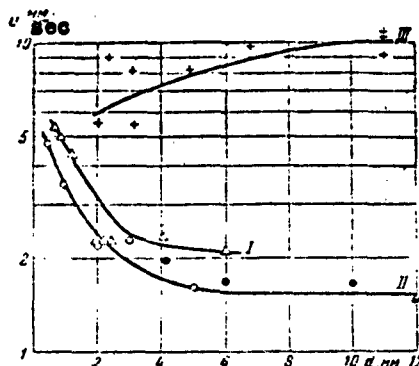


Fig. 1 The dependence of  $v$  on the thickness  $d$  of the oxidizer layer: I -- a flat layer of  $\text{KC1O}_4$  ( $\delta = 0.92$ ) between two layers of Plexiglas,  $p = 20$  atm (gage); II --  $\text{KC1O}_4$  ( $\delta = .67$ ) in flat  $\odot$  and cylindrical  $\bullet$  shells made of Plexiglas and in a steel cell with its front wall made of Plexiglas  $\ominus$ ,  $p = 10$  atm (gage); III --  $\text{BaO}_2$  ( $\delta = 0.35$ ) in a flat cell made of Plexiglas,  $p = \text{atm}$  (gage).

It is quite natural to associate the dependence of the flame speed  $v$  on the thickness  $d$  of the oxidizer layer with the mutual effect of the flames propagating along the left- and right-hand boundaries of the layer  $d$ . If effective heat exchange, increasing as  $d$  decreases, is possible between the flames, then the tip of each cavity receives heat not only from "its own", but also from the neighboring flame. In this case  $v$  increases as  $d$  decreases (until  $d$  becomes so small that the total heat generated, which decreases as the mass of the oxidizer decreases, is no longer able to compensate for the losses to the surrounding layers). It is this dependence which takes place in the case of the system Plexiglas- $\text{KC1O}_4$ -Plexiglas, where the flames are separated by a gaseous layer, and on the tip itself--by liquid  $\text{KCl}$ , which is continuously stirred by streams of gas. On the other hand, in the case of the system Plexiglas- $\text{BaO}_2$ -Plexiglas the flames

are separated by a layer of solid porous black sediment, which prevents heat exchange, and the flame speed decreases as  $d$  decreases (owing to a decrease in the total heat generated). If the sediment is removed, the flame speed increases.

The effect of the chemical nature of the components. Combustion along the surface of contact of the components depends not only on diffusion, but, to a lesser extent, on kinetic factors. The role of the latter shows up mainly in the fact that combustion cannot be achieved for anywhere near the number of combinations, which, according to calculations, produce a sufficiently great thermal effect. Thus not one of the large number of nitrates tested by us supports combustion (in contact with Plexiglas), although the calculated thermal effects of  $\text{Pb}(\text{NO}_3)_2$ ,  $\text{Ba}(\text{NO}_3)_2$ ,  $\text{Sr}(\text{NO}_3)_2$ , and especially of  $\text{LiNO}_3$ , are considerably higher than those of  $\text{BaO}_2$  or  $\text{KMnO}_4$ . This is apparently due to the fact that in the decomposition of nitrates it is not oxygen that is given off, but the relatively inert nitrogen dioxide.

We are not able to achieve combustion (in contact with  $\text{KClO}_4$ ,  $p \leq 60$  atm) with those fuels (polyethylene terephthalate, perfol', galalith, phenol-formaldehyde resin FKPM-15, and others), during the thermal decomposition of which in the presence of oxygen a large condensed residue is formed.

For those systems in which combustion can be achieved along the surface of contact the effect of the chemical nature of the components leads to considerable discrepancies in the absolute values of the flame speed and its dependence on pressure.

The effect of pressure. The dependence of the flame speed along the surface of contact of the components on the pressure of the inert gas is shown in Figs. 2 and 3. For the four oxidizing agents studied (in contact with Plexiglas) the curves  $v(p)$  differ considerably from

each other. For  $KClO_4$  the dependence is linear; for  $KClO_3$ , somewhat more strongly linear; for  $KMnO_4$   $v$  is practically independent of  $p$  in the range from 5 to 100 atm (in this case the absolute value of  $v$  is very small); finally, for  $BaO_2$  there is a maximum speed (at sufficiently high  $p$  combustion cannot take place at all with this oxidizer)\*.

The presence of a maximum for  $BaO_2$  is apparently related to the fact that an increase in pressure, on the one hand, accelerates the reaction between oxygen and the products of decomposition of the polymer (and also increases the completeness of combustion and the flame temperature), but, on the other hand, inhibits the decomposition of  $BaO_2$ , owing to the reversibility of the reaction



In the case of the fuels studied (in combination with  $KClO_4$ ) the dependence  $v(p)$  is close to linear and changes comparatively little, when we switch from one fuel to another. Only in the case of polystyrene at  $p \leq 40$  atm does  $v(p)$  turn out to be considerably steeper. It may be noted (see Table) that in the case of the thermoplastic polymers studied the value of  $v$  increases, as the softening point of the polymers (the melting point of crystalline polymers or the vitrification point of amorphous polymers) increases.

Polymer	Softening Point °C	v, mm/sec	
		p=20 atm (case)	p=40 atm (case)
Vinylplast	75	2.0	2.8
Flexiglas	100	2.8	5.0
Polyethylene	120	3.5	5.9

\* The experiments with  $BaO_2$  and  $KMnO_4$  were carried out at a low relative oxidizer density ( $\delta = 0.53$  in the case of  $KMnO_4$  and  $\delta = 0.35$  in the case of  $BaO_2$ ), since combustion could not be achieved at large  $\delta$ .

Effect of the relative density of the oxidizer. Tests were conducted on the system  $\text{KClO}_4$  + Plexiglas, where the relative density of the  $\text{KClO}_4$  varied from  $\delta = 0.5$  to  $\delta = 0.9$ . It was found (Fig. 4) that at a low flame speed (low pressures) the value of  $\underline{v}$  is practically independent of  $\delta$ , while the mass velocity  $v\delta$  decreases linearly as  $\delta$  decreases. On the other hand, at large  $\underline{v}$  (high p) the value of  $\underline{v}$  increases rapidly as  $\delta$  decreases, and the product  $v\delta = f(\delta)$  has a maximum. It is quite natural to attribute this difference to the fact that at sufficiently large  $\underline{v}$  a considerable role begins to be played by convective heat transfer from the combustion products penetrating into the fresh oxidizer. The penetration is facilitated by a decrease in  $\delta$ , but at the same time the per volume generation of heat decreases, thereby leading to the appearance of a maximum on the curve  $v\delta = f(\delta)$ . In the experiments in the other sections we tried as much as possible to eliminate the effect of convection, by increasing  $\delta$  and by placing an upper limit on the range of working pressures.

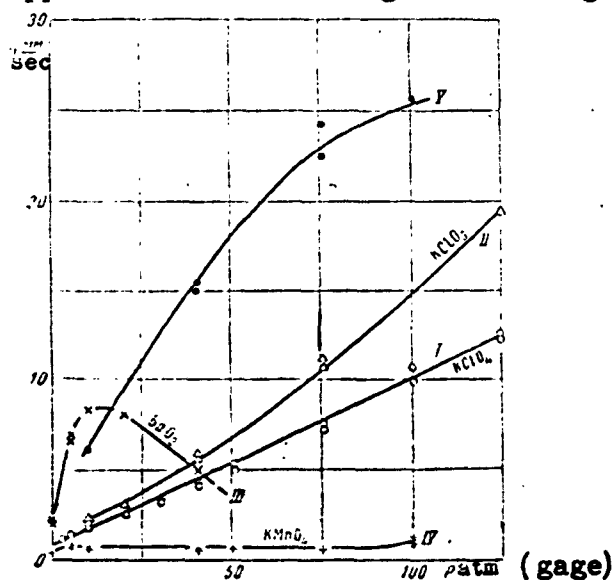


Fig. 2. The dependence  $v(p)$  for different oxidizers in a cylindrical shell ( $d = 6$  mm) made of Plexiglas: I --  $\text{KClO}_4$  ( $\delta = 0.88$ ); II --  $\text{KClO}_3$  ( $\delta = 0.88$ ); III --  $\text{BaO}_2$  ( $\delta = 0.35$ ); IV --  $\text{K}_2\text{MnO}_4$  ( $\delta = 0.55$ ); V -- stoichiometric mixture of Plexiglas and  $\text{KClO}_4$  (particle size  $\sim 10 \mu$ )

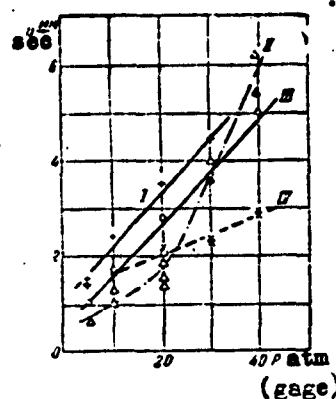


Fig. 3. The dependence  $v(p)$  for a flat layer of different fuels in contact with  $KClO_4$  ( $\delta = 0.68$ ): I -- polyethylene, II -- polystyrene, III -- Plexiglas, IV -- vinylplast

The burning of films of variable thickness applied on a thick base. In order to investigate the effect of kinetic factors on the value of  $\underline{v}$ , we studied the propagation of a flame along a flat surface of contact of  $KClO_4$  + Plexiglas, various thin (0.01-0.1 mm) films having been pressed (or applied from a solution) onto the Plexiglas surface. It was found that a polyethylene film 0.1 mm thick increases  $\underline{v}$  by 15-20%, a film of polytetrafluoroethylene (fluoroplast-4) 0.06 mm thick reduces the value of  $\underline{v}$  by almost half, a perfol' film 0.06 mm thick or a film of polyethylene terephthalate 0.15 mm completely suppresses combustion (at  $p \leq 50$  atm). Films of bitumen, natural rubber, and a number of other materials have no effect on the value of  $\underline{v}$ . In the case of polyethylene and fluoroplast-4 the dependence of the flame speed on the film thickness ( $d_f$ ) was investigated for several values of  $p$  (Fig. 5). Obviously, when  $d_f = 0$ ,  $\underline{v}$  corresponds to pure Plexiglas, but when  $d_f$  is sufficiently large,  $\underline{v}$  corresponds to the polymer out of which the film is made. In the case of polyethylene, when  $d_f$  increases, the flame speed increases smoothly, and at a certain  $(d_f)_*$  the curve

$v(d_f)$  emerges onto a plateau. The value of  $(d_f)_*$  decreases as the pressure increases and amounts to  $\sim 0.9$  mm at  $p = 10$  atm (gage) and  $\sim 0.2$  mm at  $p = 30$  atm (gage). In the case of fluoroplast-4 the dependence  $v(d_f)$  has a complex configuration. When  $d_f$  increases, the value of  $v$  at first decreases rapidly, then the curve  $v(d_f)$  emerges onto a plateau, and at a certain sufficiently large thickness (which is all the greater, the higher the pressure, and amounts to 0.25 mm at  $p = 20$  atm (gage) and to  $\sim 0.6$  mm at  $p = 30$  atm) combustion ceases\*.

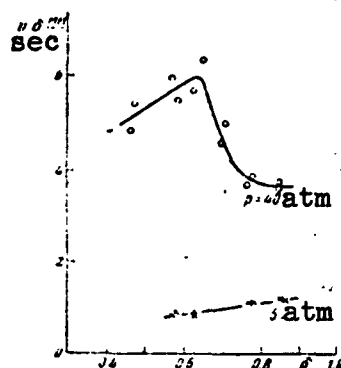


Fig. 4. The dependence of  $v\delta$  on  $\delta$  for  $KC10_4$  in a cylindrical shell made of Plexiglas

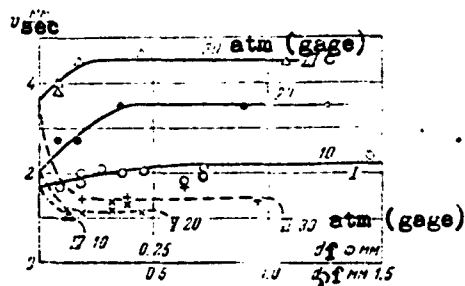


Fig. 5. The dependence of  $v$  on the thickness  $d_f$  of a film applied on a Plexiglas plate in contact with  $KC10_4$  ( $\delta = 0.68$ ): I --  $\odot$ ; II --  $\bullet$ ; III --  $\Delta$ , polyethylene film ( $d_{pf}$ ); IV --  $\Delta$ ; V --  $\times$ ; VI --  $+$ , film of fluoroplast-4 ( $d_{ff}$ )

The fuel burnup angle. We measured the shape of the cavity forming during the course of combustion in a flat Plexiglas plate (the system:

\* When  $p = 10$  atm (gage), the curve  $v(d_f)$  for fluoroplast-4 breaks off before emerging onto a plateau.

a layer of  $\text{KC10}_4$  of thickness  $d$  between two thick Plexiglas plates). The experiments were conducted at  $p = 1$  atm (abs) (in nitrogen), and at a specified moment the flame was suddenly extinguished by a stream of water. The topography of the plates was measured on a microscope with micrometric feed along three mutually perpendicular axes. Figure 6 shows the average profile\* obtained from many measurements of the cavity formed in the case of oxidizer layers 25 mm and 3.8 mm thick. The burnup angle  $\alpha$  (the angle between the tangent to the profile of the cavity at a given point and the direction of propagation of the flame) decreases smoothly, as the distance from the tip increases. On the initial section (2-3 mm)  $\alpha$  changes very little and amounts to  $\sim 32^\circ$ , when  $d = 25$  mm, and to  $\sim 13^\circ$ , when  $d = 3.8$  mm. Thus, when the flame speed increases as a result of a decrease in the thickness of the oxidizer layer, the burnup angle becomes more acute (i.e., the cavity becomes less deep).

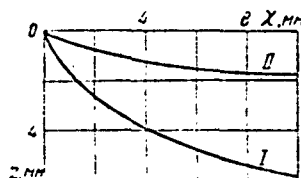


Fig. 6. Averaged profile of the cavity formed in a Plexiglas plate during combustion in contact with  $\text{KC10}_4$  ( $\delta = 0.68$ ),  $p = 1$  atm (abs): I --  $d\text{KC10}_4 = 25$  mm,  $v = 0.50$  mm/sec; II --  $d\text{KC10}_4 = 3.8$  mm,  $v = 0.97$  mm/sec.

We also measured the average burnup angle  $\alpha_{av}$  for vinylplast with  $p = 10, 20, 30$ , and  $40$  atm (gage). In the measurements we made use of the fact that during the burning of a plate made of this polymer, a plate pressed out of several thinner plates, parallel lines may be noted on the photorecorder. Of these the first (the most distinct)

\* In the plane perpendicular to the burning front and to the initial surface of the plate.

corresponds to the movement of a point of light over the surface of the first thin plate (in this case the light passes through the entire thickness of the plate and undergoes absorption on the boundaries of all the thin plates). The second line corresponds to the moment when the combustion moves out onto the surface of the second thin plate (and the absorption of light decreases abruptly) etc. Measuring with the aid of the photorecorder the distance (along the direction of propagation of the flame) between the lines and knowing the thickness of the thin plates, we obtain (taking into account the enlargement of the optical system of the photorecorder) the average value of the burnup angle. In the case of vinylplast it was shown that when the flame speed  $v$  increases as a result of an increase in pressure, the burnup angle decreases. An empirical formula appearing as follows was obtained:

$$\sin \alpha_{av} = \frac{0.1}{[v(p)]^{0.55}} \sim \frac{1}{\sqrt{v(p)}}$$

where  $\alpha_{av}$  was measured on a plate 0.9 mm thick.

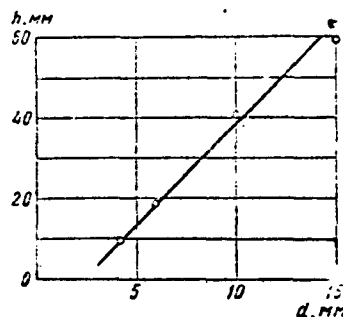


Fig. 7. The dependence of the averaged height of the nucleus of the flame jet on  $d$  during the burning of  $KClO_4$  ( $\delta = 0.68$ ) in cylindrical shells made of Plexiglas

Burning along the fuel-oxidizer boundary on a free surface. We investigated the propagation of a flame along the line Plexiglas- $KClO_4$ .



on the free (bordering on the gas) edge of a cube, one half of which consisted of  $KClO_4$ , while the other half consisted of Plexiglas. The average flame speed was found to be equal to  $\sim 7$  mm/sec, when  $p = 20$  atm (gage), and  $\sim 19$  mm/sec, when  $p = 40$  atm (gage) (the dispersion of the results of these tests is considerable). On the other hand, the flame speed along a flat surface of contact of sufficiently thick layers of Plexiglas and  $KClO_4$  is only 2.8 and 5.0 mm/sec, respectively. Thus the presence of a free surface sharply increases the flame speed. This is probably due to the appearance of a stream of hot combustion products, which determines the movement of the tip of the flame. The existence of this stream is due to the fact that the pressure near the surface of the burning condensed material is higher than the pressure far from it.

The dependence of the height of the jet on the characteristic dimension of the oxidizer layer\*. During the burning of  $KClO_4$  pressed into a thick cylindrical Plexiglas shell with an inner diameter  $d$  in an inert-gas (nitrogen) atmosphere there appears a flame jet with a fairly pronounced nucleus and a large, comparatively weakly luminescent tail with blurred contours. With the aid of a motion-picture camera the height  $h$  of the nucleus of the jet was measured for various  $d$ . In each given test the value of  $h$  fluctuated noticeably from frame to frame around a certain average value. The latter remained constant during a considerable part of the burning time for  $d = 4$  to 10 mm, but decreased for  $d = 15$  mm, as the charge was burned up. The dependence of the height of the nucleus of the jet on  $d$  was averaged over many measurements and is shown in Fig. 7. Apparently, we must assume that

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\* The experiments were performed by Yu. V. Frolov.

$h \sim d$ . The weakening of the dependence  $h(d)$ , when  $d = 15$  mm, is probably due to the fact that the smoke (consisting of finely dispersed KC1) forming during the combustion veils the apex of the nucleus of the jet all the more strongly, the larger  $d$  is.

The fact that  $h \sim d$  (and not  $d^2$ ) to a certain extent attests to the turbulent nature of the mixing.

Discussion of the results. The propagation of a flame along the surface of contact of a solid fuel and an oxidizer under the conditions studied includes the gasification of the components under the action of the heat flow from the flame; the mixing (owing to the collision of flows directed at an angle to each other and to molecular diffusion); the heating of the products of decomposition of the initial components and the combustion which follows. Moreover, near the tip combustion of a homogeneous mixture apparently takes place (the mixing manages to take place within the limits of the heating zone), and a diffusion flame sets in above.

The interpretation of the physical meaning of the flame speed  $v$  along the surface of contact (and consequently, the dependences of  $v$  on pressure, the characteristic dimension of the component layer, etc) gives rise to a number of difficulties. However, the experiments carried out enable us to draw certain partial conclusions.

It may be asserted that the flame speed significantly depends only on those processes which take place in a small zone near the tip. The size of this zone can be estimated from the following experimental data.

If the surface of a thick plate of fuel is coated with a film of variable thickness  $d_f$  of a second fuel, the value of  $v$  changes only until the thickness of the film reaches a certain value  $(d_f)_* = f(p)$ .

Hence it follows that the value of  $\underline{y}$  is affected by the processes in a triangular (in cross section) region with the catheti  $(d_f)_*$  and  $(d_f)_* \cot \alpha_{av}$  (where  $\alpha_{av}$  is the average burnup angle of the second fuel) and in the corresponding region of the oxidizer. In a steel cell, in which only the front wall is made of a thick fuel plate the flame speed  $\underline{y}$  increases as the thickness of the oxidizer layer increases (owing to a decrease in the heat losses to the steel), but only as long as  $d \leq d_*$ . This gives for the zone of influence a result analogous to the preceding one.

Certain conclusions concerning the meaning of the value of  $\underline{y}$  can be made on the basis of a measurement of the shape of the cavity which forms in the fuel plate during the course of combustion (the shape of the cavity in the oxidizer layer could not by any means be reliably measured, owing to the presence of a condensed residue). In order for the shape of the cavity to be stationary, all the points of its contour must shift through equal segments per unit of time in the direction of propagation of the combustion.

Therefore for any point of the contour

$$\frac{u}{v} = \sin \alpha \quad (1)$$

where  $\underline{u}$  is the normal rate of gasification, and  $\alpha$  is the fuel burnup angle at a given point. The normal rate of gasification is related by a simple formula to the heat flux  $q$  from the flame

$$\rho u [c(T_s - T_0) + \lambda] = q \quad (2)$$

where all the quantities pertain to a solid fuel, with  $T_s$  being the surface temperature and  $\lambda$  the heat of gasification. Since the quantities  $T_s$  and  $\lambda$  depend only slightly on  $\underline{u}$ , in the first approximation we can take

$$u \sim q \quad (2')$$

Given the profile of the cavity, we can determine from (1) and (2) the heat flux at each point. In particular, wherever the surface of the cavity becomes almost parallel to the initial surface of the plate (the burnup angle is close to zero), the heat flux also tends to zero. On the tip itself equation (1) should give either  $u_* = v$ ,  $\alpha_* = \pi/2$ , or  $u_* < v$ ,  $\alpha_* = \arcsin u_*/v$ . In the first case  $v$  represents the normal burning velocity, which is maximum with respect to all the other points of the cavity contour. In the second case  $v$  represents the rate of ignition of the fuel surface. This possibility can be readily understood in the particular case, where the decomposition of the oxidizer precedes the decomposition of the fuel, and the surface of the fuel over a certain distance in front of the tip is wholly or partially exposed.

A consideration of the processes occurring on the tip itself requires sufficiently accurate experiments and may possibly lead to results that differ for different systems. In the case of the Plexiglas-KC10<sub>4</sub> system, the one most thoroughly studied by us, the second case is probable, since the measurement lines give for  $\alpha_*$  values considerably less than  $\pi/2$  and since the quantity  $v$  is very sensitive to the application of thin films of certain polymers on a Plexiglas surface. Let us note, in conclusion, that in a number of cases it is sufficiently simple to consider the tip as a certain singular point, the velocity of motion of which  $v$  depends on the pressure  $p$ , the characteristic dimension  $d$  of the component layer, etc., differently from the normal rate of gasification over a not-too-small distance from the tip. Moreover, for the cases studied by us  $\partial v/\partial p > \partial u/\partial p$ ,  $\partial v/\partial d > \partial u/\partial d$ , etc., since the average burnup angle decreases as  $v$  increases.

Submitted on July 30, 1960

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